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(54) A METHOD FOR OBTAINING TOBACCO AROMA SUBSTANCES

(71) We, H.F. & Ph.F. REEMTSMA, a body corporate, of Parkstrasse 51, 2000 Hamburg 52, Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The invention relates to a method for obtaining tobacco aroma substances, in the course of which the tobacco is subjected to an extraction treatment.

There has been a previous proposal to extract tobacco with water and then, following this, with alcohol as well and in certain cases after combining the extracts, and reducing them in bulk, so that if required a solvent free residue may be obtained, then to apply the extracts to tobacco. The tobacco treated with the extract in this manner is intended, owing to the measures employed, to have an improved taste or to have a smoke flavour in accordance with the tobaccos used for obtaining the extract.

In accordance with the German Patent Specification 1,517,252 tobaccos, preferably in the raw condition, are extracted with the help of solvents cooled to a low temperature, for example iced water. The extract is caused to solidify and then dried by freeze drying. A light-brown product is produced with a high aroma content, which is applied to tobacco at the rate of 5 to 10% by weight in the form of a powder or in the form of a solution. The aroma of the product is intended to influence the smoke flavour.

Since the fermented tobacco which can be worked and used as a basis in the method mentioned consists of up to approximately 50% by weight of soluble components and this part must also be extracted for commercial reasons, the maximum treatment indicated of the tobacco whose smoke flavour is to be influenced, with 10% of its weight of extract powder, whose proportion of soluble components is admittedly raised by 20% of its weight, but the flavour level is only raised by the comparatively small amount of approximately 10%, and, however, the condensate content of smoke of

tobacco goods produced from tobaccos modified in this manner is consequently increased by raising the soluble tobacco components.

There is thus no substantial enhancement of the flavour level which is neutral, more especially, as regards the condensate content of the smoke.

In accordance with a further prior proposal in the form of the German Patent Specification 1,109,583 the solvent extracts from tobacco were to be reacted for example with alcohols and catalysts in order to obtain flavouring materials or substances from the tobacco. With this method it is not possible to obtain any aroma substances required for aromatising, and free from ballast substances.

An object of the invention is to provide a method with which it is possible to isolate the natural aroma substances of tobacco so as to be free as far as possible of undesired associated substances. Furthermore, the invention includes obtaining precursors of such natural aroma substances with the above-mentioned substances or alone, the aroma substances themselves being converted.

The present invention provides a method for obtaining tobacco aroma substances in the course of which the tobacco is subjected to an extraction treatment, wherein exclusively the components of the tobacco which are soluble in a solvent or solvent mixture lying between trichloroethylene and ethyl acetate inclusive in the mixotropic series of K. Macek in Handbuch der Papierchromatographie, vol. I, VEB Gustav Fischer-Verlag, Jena (1958), are extracted and the residue obtained after removing solvent or solvent mixture from the extract is subjected to heat treatment at a temperature from 30°C to 260°C.

A corresponding English reference to the German handbook mentioned above is *Paper Chromatography*, Academic Press (1969) at page 116. Unfortunately the English does not correspond exactly with the German handbook because in the English version the compound "trichloroethylene" is missing and this compound is present in the German

version between propylchloride and toluene.

5 In accordance with a further feature of the invention the tobacco extract is isolated from those substances, which are both soluble in the aforesaid solvent or solvent mixture and also in other solvents lying outside the limits set.

10 In the case of research carried out in connection with the development of the present invention it has been found surprisingly that only approximately 5 to 20% of the proportion of solid extract, corresponding to approximately 2.5 to approximately 10% of the dry weight of the tobacco in normal tobacco, is responsible for the smoke flavour of tobacco goods produced using it.

20 In this solid part of the extract there is a surprisingly high proportion of precursors of the materials that give rise to the smoke flavour, which precursors can be activated by suitable heating. They are contained in conventional tobacco without contributing to strengthening of the aroma and constitute a latent aroma reserve, which in accordance with the method in accordance with the invention are also made use of.

30 Advantageously in accordance with a further development of the invention the method is so carried out that the tobacco is extracted with a first solvent or solvent mixture which lies in the above-mentioned series of K. Macek between water and ethylacetate, the extract is substantially freed from the first solvent or solvent mixture the resulting extract residue is taken up in a second solvent, or solvent mixture, alone or mixed with water, the second solvent or solvent mixture being incompletely soluble in hydrocarbons with 5 to 12 carbon atoms but being miscible with water at least in a weight ratio of 1:25, distribution of the dissolved residue is carried out against a third solvent or solvent mixture which lies in the mixotropic series of K. Macek between toluene and liquid paraffin inclusive, the more polar of the two phases is so reduced in bulk that the residue contains at the most 8% V/V of solvents, the residue so reduced in bulk in this manner is taken up in water and the solute contained in it; distributed against a fourth solvent or a solvent mixture whose polarity is less than that of the third solvent or solvent mixture, and the dissolved material is recovered from the less polar of the two phases.

50 This residue from the less polar phase as such constitutes a product, which improves the smoke properties of the tobacco to which it is applied so as to be better than those of an untreated control tobacco. An enhancement of this improvement in quality is obtained by heating the product before the application or, in particular cases to be determined by preliminary tests, after the

application, to temperatures between 50°C and generally not more than 260°C, preferably however not more than 210°C. The time of heat treatment depends on the size of the batch and naturally also on the most favourable temperature which is to be determined. Generally the time of treatment is less when the temperature selected is higher. In the case of higher temperatures lying above 210°C, this time is rapidly shortened down to a few seconds.

70 In the case of a modified embodiment of the method in accordance with the invention the tobacco is extracted with a first solvent or solvent mixture which lies in the mixotropic series of Macek between water and acetonitrile. The extract is freed of solvent and the residue, after taking up in a second solvent or solvent mixture, which is not completely soluble in a hydrocarbon with 5 to 12 C-atoms, but is capable of being mixed with water at least in a ratio of 1:25, is distributed against a third solvent or solvent mixture, which lies in the mixotropic series of Macek between trichloroethylene and ethyl acetate. Then the residue is taken from the less polar phase. The residue as such already constitutes a product which is suitable for improving the smoke properties themselves of the same tobacco on which the extraction has been carried out, but is however capable of bringing about a further improvement of the smoke properties of tobacco treated with it, when it is subjected to a heat treatment at temperatures between 30°C and generally not above 210°C.

90 The method in accordance with the invention can also be carried out in such a manner that the tobacco is extracted with a first solvent or solvent mixture which lies in the eluotropic series of Macek between n-hexanol and liquid paraffin, the extract is freed from solvent and the residue, after being taken up in a second solvent or solvent mixture which is the eluotropic series of Macek lies between water and toluene, is distributed against a third solvent or solvent mixture which lies in the eluotropic series of Macek between toluene and liquid paraffin. The residue, which is obtained from the more polar phase possibly after previous distribution against a fourth solvent or solvent mixture, which in the eluotropic series of Macek lies between water and acetonitrile, is as such suitable for improving the smoke properties of tobacco treated with it, but leads to results with a far better flavour level, when the residue is subjected to heat treatment.

120 Reversing the process steps in the method in accordance with the invention as regards the polarity of the tobacco components or their precursors responsible for the tobacco smoke taste in substance it is also possible

in accordance with the invention to extract the tobacco with a first solvent or solvent mixture which lies in the mixotropic series of Macek between toluene and liquid paraffin and following this with a second solvent or solvent mixture, which in the mixotropic series of Macek lies between toluene and n-hexanol. This residue which is obtained from the more polar solvent or solvent mixture after evaporation is, possibly after previous taking up into a third solvent or solvent mixture, which in the mixotropic series of Macek lies between water and methanol, and distribution against the solvent or solvent mixture, which in the mixotropic series of Macek lies between n-hexanol and toluene, more particularly is suitable for improving the smoke properties of a tobacco difference to the tobacco which has been used for obtaining the extract.

A method which is particularly suitable for the purposes of the invention comprises exhaustively extracting the tobacco with an aqueous aliphatic alcohol, free in the extract from solvent by evaporation, taking up the residue in 85% methanol, distributing it against hexane and producing the methanol phase in bulk to such an extent that the residue is practically completely free of alcohol and the water containing residue is distributed between chloroform and water and the residue is obtained from the chloroform phase which has been substantially freed from solvent.

For extraction use is preferably made of fermented tobacco.

As an extraction agent or solvent it is particularly preferred to use isopropanol alone or mixed with water.

The extraction is preferably carried out between 45°C and 85°C.

The heat treatment of the residue obtained is carried out so as to produce particularly valuable products, in accordance with a further embodiment with the invention, between, conveniently, 110 and 180°C.

The heat treatment is continued in this case for up to 9 hours after achieving the treatment temperature.

It has admittedly been found to be favourable to carry out the heat treatment of the products obtained from the extracts on the products themselves, but it is basically also possible to heat the tobacco, whose smoke properties are to be influenced after the application of the product, from the extracts, which have already been subjected to the heat treatment or have not been subjected to heat treatment. In this respect the type of heat source and the means for conducting heat only play a subordinate part in the invention and more especially only influence the duration of heat treatment. Furthermore, it may also be convenient in special cases to subject the tobacco, used as a

starting material, before extraction, or the extract before further treatment to heat treatment in a suitable manner.

The following Examples illustrate the invention.

EXAMPLE 1

50 g dry tobacco of a tobacco mixture used as a basis for commercially conventional cigarettes were extracted with a mixture of 270 g pure isopropanol and 60 g water, corresponding to 330 g 82% W/W isopropanol and the solvent was distilled off azeotropically. The distillate consisted of 270 g pure isopropanol and 40 g water, corresponding to 310 g 88% W/W isopropanol. The aqueous extraction residue of 39 g, consisting of 20 g water and 19 g dry residue, corresponding to 38% by weight of the tobacco used, was taken up in 135 g pure methanol and 10 g water, corresponding to 82% W/W methanol taking into account water from the extraction residue and was distributed against 3 subsequent portions of 30 g hexane. The quantity of substance soluble in hexane amounted to 1.75 g, corresponding to 3.5% by weight of the quantity of tobacco used. The more polar phase was distilled off azeotropically and led to a distillate consisting of 135 g of pure methanol and 17 g water, corresponding to 152 g 89% W/W methanol.

The aqueous residue consisted of 13 g water and 17.25 g dry matter and was taken up in 135 g water and distributed against three subsequent portions (75 g) of chloroform. The amount of dry matter in the aqueous phase amounted to 16.0 g, corresponding to 32.0% of the dry tobacco used and that of the chloroform phase amounted to 1.25 g, corresponding to 2.5% of the quantity of dry tobacco. The solvent-free residue from the chloroform phase was heated for 5 minutes at 180°C and as a result developed a full and unusual tobacco aroma, which was far superior both quantitatively and qualitatively to that of the tobacco used and imparted to the smoke taste of the starting tobacco treated with it a full, sweet and complete flavour soundness in comparison with an untreated control tobacco.

EXAMPLE 2

32 g of conditioned tobacco of a tobacco mixture as used as a basis for ordinary commercial cigarettes with a moisture content of 15.5%, corresponding to 27 g dry tobacco were shaken up for 24 hours with 360 ml ethyl acetate, the solvent was renewed and the extraction was repeated until the solvent was colourless. The combined extracts were evaporated to dryness at 20 Torr. The yield amounted to 1.99 g of dry residue, corresponding to 7% of the dry

tobacco used, and the residue was taken up in 25 ml of 85% V/V aqueous methanol and distributed against three successive 8 ml portions of hexane. The hexane phases were substantially freed of solvent under vacuum and yielded a dry residue of 8.36 g. corresponding to 3.2% of the dry tobacco used.

The more polar phase was so reduced in volume under vacuum that the residue was practically free of methanol and only contained 1.8 g of water. After the addition of a further 20 g of water to this aqueous residue distribution was carried out with three successive portions of 8 ml of chloroform. After decanting and reducing the chloroform phase in bulk by evaporation until all solvent was removed, a residue of 0.49 g. corresponding to 1.3% of the dry tobacco used, was obtained, which after heating for 7 minutes at 165°C developed a full and intensive tobacco aroma, which as regards its qualitative properties was comparable with the product in accordance with example 1.

EXAMPLE 3

50 g of dry tobacco dust were extracted with 270 g of pure isopropanol and 60 g water, corresponding to 330 g of 82% W/W isopropanol, at 74°C and the solvent was distilled off azeotropically. The aqueous extraction residue of 29.25 g. consisting of 20 g of water and 9.25 g of dry residue, corresponding to 18.5% by weight of the dry tobacco dust used was taken up in 135 g of pure methanol and 10 g of water, corresponding to a 82% W-W methanol allowing for the water from the extraction residue and was distributed against three 30 g portions of hexane. The quantity of substance soluble in hexane amounted to 1.5 g. corresponding to 3.0% by weight of the amount of tobacco dust used. The more polar phase was distilled off azeotropically and an aqueous residue was obtained, which consisted of 13 g of water and 7.75 g of dry matter. This residue was taken up in 135 g of water and distributed against three 120 g portions of chloroform.

The amount of dry substance in the aqueous phase amounted to 2.75 g. corresponding to 5.5% by weight of the dry tobacco dust used and that of the chloroform phase amounted to 5 g. corresponding to 10% of the tobacco dust used. The practically solvent free residue from the chloroform phase was heated to 155°C and developed a full and sweet tobacco aroma which improved the smoke taste of the starting tobacco treated with it as compared with untreated tobacco as regards its intensity while increasing the expression of the tobacco.

It is naturally possible within the scope of the invention to process the aroma sub-

stances obtained, possibly using inflammable, ash-forming and/or substances influencing combustion and/or the smoke taste to produce products suitable for processing in tobacco goods production. It is thus possible with the tobacco aroma materials in accordance with the invention to charge carriers which as such are neutral as well so that in this manner a substance for smoking is produced which is only defined by the aroma materials.

WHAT WE CLAIM IS:—

1. A method for obtaining tobacco aroma substances in the course of which the tobacco is subjected to an extraction treatment, wherein exclusively the components of the tobacco which are soluble in a solvent or solvent mixture lying between trichloroethylene and ethyl acetate inclusive in the mixotropic series of K. Macek in *Handbuch der Papierchromatographie*, vol. J, VEB Gustav Fischer-Verlag, Jena (1958), are extracted and the residue obtained after removing solvent or solvent mixture from the extract is subjected to heat treatment at a temperature from 30°C to 260°C.

2. A method as claimed in claim 1, in which the tobacco extract is separated from those materials present in the tobacco which are soluble not only in a solvent or solvent or solvent mixture within the specified range but also in solvents lying outside the range.

3. A method as claimed in claim 1, in which the tobacco is extracted with a first solvent or solvent mixture which lies between water and ethyl acetate inclusive in the mixotropic series of Macek, the extract is substantially freed from the first solvent or solvent mixture, the resulting extract residue is taken up in a second solvent or solvent mixture, alone or mixed with water, the second solvent or solvent mixture being incompletely soluble in hydrocarbons with 5 to 12 C-atoms but being miscible with water at least in a weight ratio of 1:25, distribution of the dissolved residue is carried out against a third solvent or solvent mixture which lies in the mixotropic series of Macek between toluene and liquid paraffin inclusive, the more polar of the two phases is so reduced in bulk that the residue contains at the most 8% of solvent, the residue reduced in bulk in this manner is taken up in water and the solute contained in it distributed against a fourth solvent or solvent mixture whose polarity is less than that of the third solvent or solvent mixture, and the dissolved material is recovered from the less polar of the two phases.

4. A method as claimed in claim 1, in which the tobacco is extracted with a first solvent or solvent mixture which lies in the mixotropic series of Macek between water and acetonitrile inclusive, the extract is

freed from solvent, and the resulting extract residue, after being taken up in a second solvent or solvent mixture which is incompletely soluble in hydrocarbons with 5 to 12 C-atoms, but is miscible with water at least in a weight ratio of 1:25, is distributed against a third solvent or solvent mixture which lies in the mixotropic series of Macek between trichloroethylene and ethyl acetate inclusive and the dissolved material is recovered from the less polar of the two phases.

5. A method as claimed in claim 1, in which the tobacco is extracted with a first solvent or solvent mixture which lies in the mixotropic series of Macek between n-hexanol and liquid paraffin inclusive, the extract is freed from said first solvent or solvent mixture, the resulting extract residue, after being taken up in a second solvent or solvent mixture, which lies in the mixotropic series of Macek between water and toluene inclusive, is distributed against a third solvent or solvent mixture which in the mixotropic series of Macek lies between toluene and liquid paraffin inclusive, the dissolved material is separated from the less polar of the two phases and optionally taken up in a fourth solvent or solvent mixture which lies in the mixotropic series of Macek between n-hexanol and trichloroethylene inclusive, distribution against a fifth solvent or solvent mixture which lies in the mixotropic series of Macek between water and acetonitrile inclusive, and separated from the less polar of the two phases prior to recovery.

6. A method as claimed in claim 1, in which the tobacco is extracted firstly with a first solvent or solvent mixture which lies in the mixotropic series of Macek between toluene and liquid paraffin inclusive, and then with a second solvent or solvent mixture which lies in the mixotropic series of Macek between toluene and n-hexanol inclusive, and the dissolved material is separated from the more polar of the two solvents or solvent mixtures, optionally after that solvent or solvent mixture has been distributed against a third solvent or solvent

mixture which in the mixotropic series of Macek lies between water and acetonitrile inclusive, and the more polar of the two last-mentioned solvents or solvent mixtures has been rejected.

7. A method as claimed in claim 1 or 2, in which the tobacco is exhaustively extracted with an aqueous aliphatic alcohol, the extract is freed from the aqueous aliphatic alcohol by evaporation, the resulting evaporation residue is taken up in 85% aqueous methanol and distributed against hexane, the methanol phase is so reduced in bulk that the residue is practically completely free from methanol, the aqueous residue from said reduction in bulk is distributed between chloroform and water and the dissolved material is recovered from the chloroform phase substantially freed of solvent.

8. A method as claimed in any of the preceding claims, in which fermented tobacco is used as starting material.

9. A method as claimed in claim 3 or claim 7, in which isopropanol alone or mixed with water is used as an extraction solvent.

10. A method as claimed in any preceding claim, in which the extraction is carried out at temperatures from 45 to 85°C.

11. A method as claimed in any preceding claim, in which the heat treatment of the residue obtained is carried out at a temperature from 110 to 180°C.

12. A method as claimed in claim 11, in which the heat treatment is carried out for up to 9 hours after reaching the treatment temperature.

13. A method as claimed in claim 1 and substantially as described above in any of the foregoing Examples.

14. A tobacco extract when produced by a method as claimed in any preceding claim.

15. A tobacco treated with an extract as claimed in claim 14.

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COMPLETE SPECIFICATION

I SHEET

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